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SUBSTITUTE SPECIFICATION**
(Submitted with Amendment B Under Rule 116)

Serial No. 09/739,477

Docket No. NEC PF-2727

MARKED SUBSTITUTE SPECIFICATIONSubmitted with Amendment B Under Rule 116

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METHOD OF PROCESSING A NANOTUBE USING A SELECTIVE SOLID STATE REACTION

5

BACKGROUND OF THE INVENTION

The present invention relates to a method of processing a nanotube, and
10 more particularly to a method of processing a nanotube, which is suitable for cutting
the nanotube and for forming a top of the nanotube.

A ~~single-layer-winded~~ single-walled carbon nanotube has an extremely
fine structure ~~[[in]]~~ of nanometer order. Properties of the ~~single-layer-winded~~
single-walled carbon nanotube have been on the investigation. The research and
15 developments of the ~~single-layer-winded~~ single-walled carbon nanotube for
application have been active. The ~~single-layer-winded~~ single-walled carbon nanotube
comprises a ~~eylindrically-winded~~ single layer of graphite hexagonal network. An
electron structure largely varies depending upon a diameter of the tube and a chiral
angle, for which reason the electrical conductivity of the carbon nanotube varies
20 between a conductivity of a metal and a conductivity of a semiconductor, and further
the carbon nanotube exhibits a property similar to one-dimensional electric
conduction.

The carbon nanotube may be applied to a field emitter. This field emitter
has been known and is disclosed in (1) W.A. de Heer, A.Chatelain,

Serial No. 09/739,477

Docket No. NEC PF-2727

MARKED SUBSTITUTE SPECIFICATIONSubmitted with Amendment B Under Rule 116

and D. Ugarte, Science 270, 1179 (1995); (2) A.G. Rinzler, J.H. Hafner, P. Nikolaev, L. Lou, S.G. Kim, D. Tomanek, P. Nordlander, D.T. Colbert, and R.E. Smalley, Science, 269, 1550 (1995); (3) P.G. Collins and A. Zettl, Appl. Phys. Lett., 69, 1969 (1996); (4) Q.H. Wang, T.D. Corrigan, J.Y Dai, P. R. H. Chang, and A.R. Krauss, 5 Appl. Phys. Lett., 70, 3308 (1997); (5) Y. Saito, K. Hamaguchi, T. Nishino, K. Hata, K. Tohji, A. Kasuya, and Y. Nishina, Jpn. J. Appl. Phys., 36, L1340 (1997); (6) J-M. Bonard, J-P. Salvetat, T. Stockli, W.A. de Heer, L. Forro, and A. Chatelain, Appl. Phys. Lett., 73, 918 (1998).

The sharpness of the top of the field emitter of the carbon nanotube
10 emphasizes the field effect and realizes the good characteristic of the field emission.

No practicable method of operating the nanotube tip and controlling the same has been established, for which reason the conventional carbon nanotube is extremely low in probability of having an optimum directionality and serving as
[[an]] a useful electron emitter. As shown in (7) Y Zhang and S. Iijima, Philos. Mag.
15 Lett., 78, 139 (1998), the most of the manufactured ~~single-layer-winded~~ single-walled carbon nanotube is large in aspect ratio or is slender and further is curved, for which reason it is difficult to practice the ~~single-layer-winded~~ single-walled carbon nanotube.

It is actually difficult that the plural nanotube tips are arranged in the same
20 direction and the plural nanotubes are aligned along a single line. As shown in the above literature (7), it is actually difficult to evaluate the nanotube tip with a probe.

Serial No. 09/739,477

Docket No. NEC PF-2727

MARKED SUBSTITUTE SPECIFICATIONSubmitted with Amendment B Under Rule 116

In order to have solved the above problem, there was proposed a method of cutting the single carbon nanotube by an oxidation using ~~[[a]]~~ nitric acid, an acid ~~mixing mixed~~ with ~~[[a]]~~ nitric acid or ~~[[a]]~~ sulfuric acid. The cutting method is disclosed in (8) K.B. Shelimov, R.O. Esenaliev, A.G. Rinzler, C.B. Huffman, and
5 R.E. Smalley, Chem. Phys. Lett., 282, 429 (1998); (9) J. Liu, M. J. Casavant, M.Cox, D.A. Walters, P. Boul, W. Lu, A.J. Rimberg, K.A. Smith, D.T. Colbert, and R.E. Smalley, Chem. Phys. Lett., 303, 125 (1999); (10) Z. Shi, Y. Lian, F. Liao, X. Zhou, Z. Gu, Y. Zhang, and S. Iijima, Solid State Comm., 112 (1999) 35.

The above cutting method for cutting the ~~single-layer-winded~~
10 ~~single-walled~~ carbon nanotube is incapable of cutting, at a specified site, the ~~single-layer-winded~~ ~~single-walled~~ carbon nanotube. Namely, the above cutting method is unable to specify the cutting site of the ~~single-layer-winded~~ ~~single-walled~~ carbon nanotube. Since the above cutting method is the chemical wet process using ~~[[the]]~~ acid such as ~~[[the]]~~ nitric acid and ~~[[the]]~~ sulfuric acid, the acid provides
15 undesirable influence to the manufacturing process. The above conventional cutting method is unsuitable for forming the micro device.

In the above circumstances, it had been required to develop a novel method of processing a nanotube free from the above problem.

20

SUMMARY OF THE INVENTION

Accordingly, it is an object of the present invention to provide a novel method of processing a nanotube free from the above problems.

Serial No. 09/739,477

Docket No. NEC PF-2727

MARKED SUBSTITUTE SPECIFICATIONSubmitted with Amendment B Under Rule 116

It is a further object of the present invention to provide a novel method of processing a nanotube without using any chemical wet process.

It is a still further object of the present invention to provide a novel method of selectively processing a top portion of a nanotube in a specific shape.

5 It is yet a further object of the present invention to provide a novel method of selectively processing a top portion of a nanotube in a specific shape which is suitable for an electron device such as a field emission electron gun.

The present invention provides a method of processing a nanotube, comprising the steps of: causing a selective solid state reaction between a selected
10 part of a nanotube and a reactive substance substrate to have the selected part only become a reaction product; and separating the nanotube from the reaction product to define an end of the nanotube.

The above and other objects, features and advantages of the present invention will be apparent from the following descriptions.

15

BRIEF DESCRIPTION OF THE DRAWINGS

Preferred embodiments according to the present invention will be described in detail with reference to the accompanying drawings.

FIGS. 1A through 1C are fragmentary cross sectional elevation views
20 illustrative of a novel method of forming a top of the ~~single-layer-winded~~ single-walled carbon nanotube in a preferred embodiment in accordance with the present invention.

Serial No. 09/739,477

Docket No. NEC PF-2727

MARKED SUBSTITUTE SPECIFICATION**Submitted with Amendment B Under Rule 116**

FIG. 2 is a view of an image of the top portions of the ~~single-layer-winded~~ single-walled carbon nanotubes taken by the transmission electron microscope in a preferred embodiment in accordance with the present invention.

5

DISCLOSURE OF THE INVENTION

The present invention in one aspect provides a method of processing a nanotube, comprising the steps of: causing a selective solid state reaction between a selected part of a nanotube and a reactive ~~substance~~ substrate to have the selected part only become a reaction product ; and separating the nanotube from the reaction

10 product to define an end of the nanotube.

It is preferable that the step of causing a selective solid-state reaction further comprises the steps of : selectively contacting the part of the nanotube with the reactive ~~substance~~ substrate ; and causing the selective solid state reaction on a contacting region of the selected part of the nanotube and the reactive ~~substance~~

15 substrate to have the selected part only become the reaction product, wherein a boundary between the reaction product and the nanotube is self-aligned to an edge portion of the contacting region of the selected part of the nanotube and the reactive ~~substance~~ substrate.

~~It is further preferable that~~ In a preferred embodiment the solid state

20 reaction is caused by heating the reactive ~~substance~~ substrate.

Serial No. 09/739,477

Docket No. NEC PF-2727

MARKED SUBSTITUTE SPECIFICATIONSubmitted with Amendment B Under Rule 116

It is ~~further more~~ preferable that Preferably the reactive ~~substance~~
substrate is heated by an irradiation of a heat ray onto the reactive ~~substance~~
substrate.

It is ~~still further~~ preferable that In such preferred embodiment the heat ray
5 is an infrared ray.

It is ~~also~~ preferable that In another preferred embodiment the reactive
~~substance~~ substrate is heated by applying a current between the reactive ~~substance~~
substrate and the nanotube.

It is also preferable that the step of contacting the part of the nanotube with
10 the reactive ~~substance~~ substrate further comprises the steps of: dispersing a carbon
nanotube into an organic solvent to form a dispersion liquid ; applying the dispersion
liquid onto a surface of the reactive ~~substance~~ substrate ; and evaporating the organic
solvent from the dispersion liquid to leave the nanotube on the reactive ~~substance~~
substrate.

15 It is ~~also~~ preferable that In a preferred embodiment the nanotube is
separated from the reaction product by rapidly cooling the reaction product.

It is ~~also~~ preferable that In one preferred embodiment the nanotube is a
~~single-layer-winded~~ single-walled nanotube.

It is ~~also~~ preferable that In another preferred embodiment the nanotube is a
20 ~~multi-layer-winded~~ multi-walled nanotube.

Serial No. 09/739,477

Docket No. NEC PF-2727

MARKED SUBSTITUTE SPECIFICATION**Submitted with Amendment B Under Rule 116**

~~It is also preferable that~~ In one preferred embodiment the nanotube is a carbon nanotube.

~~It is also preferable that~~ In another preferred embodiment the nanotube is a boron nitride based nanotube.

5 It is also preferable that the reactive substance substrate is a metal.

~~It is further preferable that~~ In one preferred embodiment the reactive ~~substance~~ substrate is Nb.

~~It is also preferable that~~ In another preferred embodiment the reactive ~~substance~~ substrate is a semiconductor.

10 ~~It is further preferable that~~ Preferably the reactive ~~substance~~ substrate is Si.

It is also preferable that the reactive ~~substance~~ substrate is in a solid state.

It is further preferable that the reactive ~~substance~~ substrate comprises a substrate having an edge.

15 It is further more preferable that the edge is defined by a hole formed in the substrate.

It is also preferable that the end of the nanotube is a top of the nanotube.

~~The second~~ In another aspect the present invention provides a method of forming a top of a carbon nanotube, comprising the steps of : selectively contacting a
20 selected part of a nanotube with a solid state reactive ~~substance~~ substrate having an edge ; carrying out a heat treatment to the solid state reactive ~~substance~~ substrate to

Serial No. 09/739,477

Docket No. NEC PF-2727

MARKED SUBSTITUTE SPECIFICATION**Submitted with Amendment B Under Rule 116**

cause a selective solid state reaction on a contacting region of the selected part of the nanotube and the solid state reactive ~~substance~~ substrate to have the selected part only become a reaction product, wherein a boundary between the reaction product and the nanotube is self-aligned to the edge of the solid state reactive ~~substance~~ substrate ;
5 and separating the nanotube from the reaction product to define a top of the nanotube.

~~It is also preferable that~~ In a preferred embodiment the solid state reactive ~~substance~~ substrate is heated by an irradiation of a heat ray onto the solid state reactive ~~substance~~ substrate.

~~It is further preferable that~~ Preferably the heat ray is an infrared ray.

10 ~~It is also preferable that~~ In a preferred embodiment the solid state reactive ~~substance~~ substrate is heated by applying a current between the solid state reactive ~~substance~~ substrate and the nanotube.

It is also preferable that the step of contacting the selected part of the nanotube with the reactive ~~substance~~ substrate further comprises the steps of
15 dispersing the nanotube into an organic solvent to form a dispersion liquid; applying the dispersion liquid onto a surface of the solid state reactive ~~substance~~ substrate ; and evaporating the organic solvent from the dispersion liquid to leave the nanotube on the solid state reactive ~~substance~~ substrate.

It is also preferable that the nanotube is separated from the reaction
20 product by rapidly cooling the reaction product.

Serial No. 09/739,477

Docket No. NEC PF-2727

MARKED SUBSTITUTE SPECIFICATIONSubmitted with Amendment B Under Rule 116

~~It is also preferable that~~ In one preferred embodiment the nanotube is a ~~single layer winded~~ single-walled nanotube.

~~It is also preferable that~~ In another preferred embodiment the nanotube is a ~~multi layer winded~~ multi-walled nanotube.

5 ~~It is also preferable that~~ In one preferred the nanotube is a carbon nanotube.

~~It is also preferable that~~ In another preferred embodiment the nanotube is a boron nitride based nanotube.

10 It is also preferable that the solid state reactive ~~substance~~ substrate is a metal.

~~It is also preferable that~~ In one preferred embodiment the solid state reactive ~~substance~~ substrate is Nb.

~~It is also preferable that~~ In another preferred embodiment the solid state reactive ~~substance~~ substrate is a semiconductor.

15 ~~It is also preferable that~~ Preferably the solid state reactive ~~substance~~ substrate is Si.

PREFERRED EMBODIMENT

20 A first embodiment according to the present invention will be described in detail with reference to the drawings. The present invention is applied to process a top portion of a ~~single layer winded~~ single-walled carbon nanotube. The ~~single layer winded~~ single-walled carbon nanotube is cut by use of Nb as a first

Serial No. 09/739,477

Docket No. NEC PF-2727

MARKED SUBSTITUTE SPECIFICATIONSubmitted with Amendment B Under Rule 116

~~substance~~ substrate to form a top of the ~~single-layer-winded~~ single-walled carbon nanotube. FIGS. 1A through 1C are fragmentary cross sectional elevation views illustrative of a novel method of forming a top of the ~~single-layer-winded~~ single-walled carbon nanotube in a preferred embodiment in accordance with the present invention.

With reference to FIG. 1A, a metal substrate, for example, a thin Nb substrate 1 was prepared as a substance which reacts with the ~~single-layer-winded~~ single-walled carbon nanotube. The Nb substrate 1 has a thickness of 2 millimeters. A hole 1a having a predetermined shape was formed ~~[[by]]~~ at a predetermined position of the Nb substrate 1 by use of an ion milling method. Further, ~~single-layer-winded~~ single-walled carbon nanotubes 2 were prepared by a laser ablation method.

The ~~single-layer-winded~~ single-walled carbon nanotubes 2 were dispersed by an ultrasonic wave into an organic solvent such as ethanol, 2-propanol and acetone to form a ~~single-layer-winded~~ single-walled carbon nanotube dispersed liquid. This dispersed liquid is applied on an upper surface of the Nb substrate 1. An organic solvent is evaporated from the dispersed liquid. In the organic solvent, ~~almost~~ most of the ~~single-layer-winded~~ single-walled carbon nanotubes 2 extend as single straight lines or are curved or form a bundle. Namely, the ~~single-layer-winded~~ single-walled carbon nanotubes 2 form a three-dimensional structure in the organic solvent. After the organic solvent was evaporated, then the ~~single-layer-winded~~ single-walled carbon nanotubes 2 are kept to form the three-dimensional structure over the Nb

Serial No. 09/739,477

Docket No. NEC PF-2727

MARKED SUBSTITUTE SPECIFICATIONSubmitted with Amendment B Under Rule 116

substrate 1. The ~~single-layer-winded~~ single-walled carbon nanotubes 2 are in contact partially with the Nb substrate 1 at the edge of the hole 1a.

The Nb substrate 1 with the ~~single-layer-winded~~ single-walled carbon nanotubes 2 was placed on a heat stage in a vacuum chamber of ~~[[a]]~~ an ultra high vacuum transmission electron microscope (UHV-TEM, JEM-2000FXVII). The vacuum chamber was sealed and vacuumed to reach a vacuum degree in the range of 1.33E-7 Pa to 1.33E-6 Pa (1E-9 Torr to 1E-8 Torr). It is possible that the vacuum chamber is filled with an inert gas such as Ar gas or N2 gas at a low pressure to form an inert gas atmosphere in the vacuum chamber.

With reference to FIG. 1B, this vacuum was still kept and an infrared ray lamp was used to irradiate an infrared ray onto the Nb substrate 1 to heat the Nb substrate 1 up to a temperature which is sufficiently high for causing a solid state reaction between the Nb substrate 1 and the ~~single-layer-winded~~ single-walled carbon nanotubes 2. For example, the highest temperature of the Nb substrate 1 may be in the range of 800-1000°C. The heat time may be in the range of 50-60 minutes.

The solid state reaction appeared on the contact region between the Nb substrate 1 and the ~~single-layer-winded~~ single-walled carbon nanotubes 2, whereby the contact portion of the ~~single-layer-winded~~ single-walled carbon nanotubes 2 contacting with the Nb substrate 1 became a reaction product 3 of NbC, wherein a boundary between the NbC reaction product 3 and the remaining ~~single-layer-winded~~

Serial No. 09/739,477

Docket No. NEC PF-2727

MARKED SUBSTITUTE SPECIFICATIONSubmitted with Amendment B Under Rule 116

single-walled carbon nanotubes 2 was aligned to the edge of the hole 1a of the Nb substrate 1.

With reference to FIG. 1C, the irradiation of the infrared ray lamp was discontinued and subsequently the sample was rapidly cooled, whereby a crack was
5 formed at the boundary between the NbC reaction product 3 and the remaining ~~single-layer-winded~~ single-walled carbon nanotubes 2, and thus the ~~single-layer-winded~~ single-walled carbon nanotubes 2 were separated from the NbC reaction product 3. As a result, the tops of the ~~single-layer-winded~~ single-walled carbon nanotubes 2 were formed.

10 FIG. 2 is a view of an image of the top portions of the ~~single-layer-winded~~ single-walled carbon nanotubes 2 taken by the transmission electron microscope. From FIG. 2, it can be understood that the top portions of the ~~single-layer-winded~~ single-walled carbon nanotubes 2 were selectively formed.

In accordance with the novel method of processing the nanotube of the
15 present invention, the solid state reaction is caused on the contact regions of the ~~single-layer-winded~~ single-walled carbon nanotubes 2 contacting with the Nb substrate 1 to cause the contact regions of the ~~single-layer-winded~~ single-walled carbon nanotubes 2 on the Nb substrate 1 become the NbC reaction product, whereby the boundary between the NbC reaction product and the ~~single-layer-winded~~
20 single-walled carbon nanotubes 2 is aligned to the edge of the Nb substrate 1. The boundary of the ~~single-layer-winded~~ single-walled carbon nanotubes 2 from the NbC

Serial No. 09/739,477

Docket No. NEC PF-2727

MARKED SUBSTITUTE SPECIFICATIONSubmitted with Amendment B Under Rule 116

reaction product define the tops of the ~~single-layer-winded~~ single-walled carbon
nanotubes 2. Non-contact portions of the ~~single-layer-winded~~ single-walled carbon
nanotubes 2 structurally remain unchanged. Namely, the tops of the ~~single-layer~~
~~winded~~ single-walled carbon nanotubes 2 are highly accurately formed without
5 changing the structures of the ~~single-layer-winded~~ single-walled carbon nanotubes 2.

The above preferred embodiment may be modified as follows. In the
above embodiment, the Nb substrate 1 was used as the substance which reacts with
the ~~single-layer-winded~~ single-walled carbon nanotubes 2. Other metals than Nb and
semiconductors such as silicon may be available as the substance which reacts with
10 the single layer winded carbon nanotubes 2.

In the above embodiment, the ~~single-layer-winded~~ single-walled carbon
nanotubes 2 were used as the nanotubes. Other nanotubes such as boron nitride based
nanotubes ~~are also available~~ may be used.

Not only a ~~single-layer-winded~~ single-walled nanotube but also
15 ~~multilayers winded nanotube are available~~ multi-walled nanotubes may be used.

In place of the infrared ray irradiation, other heating methods such as a
resistance heating method of applying a current between the ~~single-layer-winded~~
single-walled carbon nanotubes 2 and the Nb substrate 1a may be ~~available~~ used.

Whereas modifications of the present invention will be apparent to a
20 person having ordinary skill in the art, to which the invention pertains, it is to be
understood that embodiments as shown and described by way of illustrations are by

Serial No. 09/739,477

Docket No. NEC PF-2727

MARKED SUBSTITUTE SPECIFICATIONSubmitted with Amendment B Under Rule 116

no means intended to be considered in a limiting sense. Accordingly, it is to be intended to cover by claims all modifications which fall within the spirit and scope of the present invention.